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Patent

**In the United States Patent and Trademark Office**

In the Application of:

**Che-Hsiung Hsu et al.**Case No.: **PE0688USNA**Application No.: **10/669,494**Group Art Unit: **1751**Filed: **September 24, 2003**Examiner: **Mark T. Kopec**For: **Water Dispersible Polythiophenes Made With Polymeric Acid Colloids**Mail Stop Amendment  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450**CERTIFICATE OF FACSIMILE TRANSMISSION**DATE: February 12, 2008

I hereby certify that this paper and six (6) pages of laboratory notebook page copies referenced herein are being facsimile transmitted to the Patent and Trademark Office to facsimile number 571-273-8300 on the date listed above.

**Declaration**  
**37 C.F.R. § 1.131**  
John H. Lanuning  
Registration No. 34,857

Che-Hsiung Hsu, Yong Cao, Daniel David LeCloux, Sunghan Kim, and Chi Zhang  
declare as follows:

1. Dr. Hsu: I received the degree of Doctor of Philosophy from the Department of Chemistry of Temple University in 1974.

Dr. Cao: I received the degree of Doctor of Philosophy from the Department of Chemistry of Tokyo University in 1987.

Dr. LeCloux: I received the degree of Bachelor of Science in Chemistry from the University of Minnesota in 1993 and the degree of Doctor of Philosophy in Inorganic Chemistry from the Massachusetts Institute of Technology in 1998.

Dr. Kim: I received the degree of Bachelor of Science from Yonsei University in 1991 and the degree of Doctor of Philosophy from Korean Advanced Institute of Science and Technology in 1996.

Dr. Zhang: I received the degree of Bachelor of Science in Polymer Chemistry from Wuhan University in 1983 and the degree of Doctor of Philosophy in Polymer Chemistry from the Institute of Chemistry, Academia Sinica, in 1989.

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2. Dr. Hsu: I am Research Fellow for E. I. du Pont de Nemours and Company, having joined DuPont in September 1981 with continuous employment with the company since that date.

Dr. Cao: I joined Uniax, Inc., the predecessor of DuPont Displays, Inc. in January 1990 and was continuously employed by Uniax and then DuPont Displays until September 2006, when I resigned, holding the title of DuPont Displays Fellow. I am currently Professor in the College of Materials Science at South China University of Technology, Guangzhou, P.R. China 510640.

Dr. LeCloux: I joined E. I. du Pont de Nemours and Company in November 1998 and have been employed continuously since then by DuPont. My current position title is Research Manager.

Dr. Kim: I joined DuPont Displays, Inc. (Santa Barbara, California) in December 2001 and have been continuously employed by DuPont Displays since then. My current position is Senior Research Scientist.

Dr. Zhang: I am employed by DuPont Displays, Inc. as Senior Research Scientist, having joined Uniax, Inc. in October 1993 and remained continuously employed by Uniax and DuPont Displays since that time. Uniax is the corporate predecessor of DuPont Displays.

3. I am a co-inventor of the subject matter claimed in the above referenced patent application.

4. I have reviewed the Office Action dated September 12, 2007. Specifically, I have reviewed the Examiner's statements on pages 3 and 4 of the Action concerning the rejection of claims 1, 8-10, 54, and 58-60 of the application as being anticipated by United States Patent Publication No. 2004/0010115 A1, filed July 11, 2002, naming Gregory Allen Sotzing as inventor.

5. The attached Exhibit contains photocopy reproductions [with dates redacted] of pages 24-25 and 35-37 of Book E100074 and page 51 of Book E104328, both of which are laboratory notebooks kept by co-inventor Che-Hsiung Hsu as corporate records of E. I. du Pont de Nemours and Company in the regular course of research according to company policy.

6. The experimental work described in the Exhibit pages was performed in laboratories at DuPont's Experimental Station in Wilmington, Delaware, U.S.A. under the direction and supervision of Che-Hsiung Hsu. All work was performed, and all laboratory records whose copies are attached were compiled, prior to July 11, 2002.

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7. The entries of pages 24-25 of Book E100074 describe the polymerization of ethylenedioxythiophene in the presence of Nafion® and related observations.

8. The entries on pages 35-37 of Book E100074 describe experimental procedures and observations. Specifically, these laboratory records depict the polymerization of ethylenedioxythiophene in the presence of Nafion® which corresponds to Example 1 in the present application, which in turn corresponds to Invention Example 1 in Provisional Application Serial No. 60/413,202 filed on September 24, 2002.

9. The entries on page 51 of Book E104328 describe experimental procedures and observations testing the conductivity and emissivity of film made from an aliquot of the PEDOT/Nafion® dispersion. The procedures confirmed operating voltage, light emission efficiency, and stress half-life of films. These entries also correspond to data presented in Example 1 in the present application and Invention Example 1 in the provisional application, with the following clarifications.

a. The data at page 51, Book E104328, lines 7-8 giving quantities of 53 g of Lewatit® S100 and 51 g of Lewatit® MP62® WS was entered there as a result of a transcription error. These quantities are correct as given for Comparative Example 1, at page 23, lines 10-11 of the application.

b. The quantities presented in Example 1, page 25, lines 24-25 showing 7.75 g of Lewatit® S100 and 7.8 g of Lewatit® MP62® WS are correct as given for Example 1.

10. The conductivity testing was performed by Chi Zhang who acquired the conductivity data appearing in Example 1 of the application (page 26, lines 6-7) and Invention Example 1 of the provisional application. This testing was also performed prior to July 11, 2002.

I DECLARE THAT ALL STATEMENTS MADE HEREIN OF MY OWN KNOWLEDGE ARE TRUE AND THAT ALL STATEMENTS MADE ON INFORMATION AND BELIEF ARE BELIEVED TO BE TRUE; AND FURTHER, THAT THESE STATEMENTS WERE MADE WITH THE KNOWLEDGE THAT WILLFUL FALSE STATEMENTS AND THE LIKE SO MADE ARE PUNISHABLE BY FINE OR IMPRISONMENT, OR BOTH, UNDER SECTION 1001 OF TITLE 18 OF THE UNITED STATES CODE.

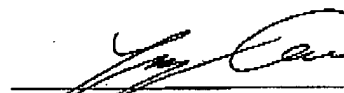
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Respectfully submitted,



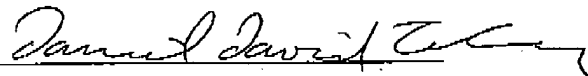
Che-Hsiung Hsu

Date: Feb. 8, 2008



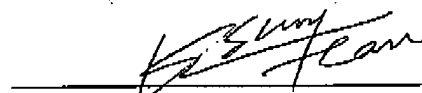
Yong Cao

Date: Feb. 3, 2008



Daniel David LeCloux

Date: Feb 11, 2008



Sunghan Kim

Date: 2/6/08



Chi Zhang

Date: 2/1/2008

TITLE

DATE

E 100074- 24.

PURPOSE

*Polymerization of Baytron - M in Nafion SE-10072*

E100074-24.

Differences: This reaction is conducted like E100061-112 except that Nafion Dispersion SE-10072 is used.

Into a corresponding 500mL Nalgene® Plastic Bottle SE-10072\* and the listed amount of DI Water were massed. The Sodium Persulfate\*\* and the aliquot of a Stock Solution of Iron(III) Sulfate\*\*\* were then massed into the bottle. The cap was then replaced and the bottle was shaken. The bottle contents were then poured into the Reaction Vessel\*. (Some losses during transfer had occurred.) Once poured into the Reaction Vessel, no gel particles were noticed. This stirred vigorously for approximately 15 minutes before being test filtered by Che®. The Reaction Mixture was then allowed to stir for another 15 minutes prior to addition of the Baytron-M®. The reaction mixture had a slight color. The Reaction Mixture temperature was 22.6°C when the Baytron-M was added. The table below shows all of the Quantities of reagents used. This table also shows the results of necessary calculations.

## NECESSARY DATA:

## Nafion

990 g/mole of Monomer Unit of Acid  
11.3 % Solid Nafion

## Sodium Persulfate

238.10 m.w. of Sodium Persulfate

## Iron(III) Sulfate

399.87 m.w. of Iron (III) Sulfate

## Stock Solution of Iron (III) Sulfate

0.0975 g Iron (III) Sulfate

18.1253 g Diluted to w/ Water

0.5098 mass % of Iron (III) Sulfate

## Baytron M

142.17 g/mole

1.334 g/mL

## Amount of Solid Sodium Persulfate Used:

1.72 g

7.224 mmoles

## Amount of Iron(III) Sulfate Used:

1.47 g of Iron(III) Sulfate Solution

0.00749 g of Solid Iron(III) Sulfate

0.0167 mmoles of Iron(III) Sulfate Used:

## Baytron M Addition:

0.630 mL of Baytron M, E100061-37

0.840 g of Baytron M (Calculated)

5.911 mmoles Baytron M (Calculated)

## Mass of Total Reaction Mixture:

319.94 g (Calculated)

## AMOUNTS ACTUALLY USED IN INITIAL REACTION MIXTURE:

## Amount of Nafion Solution Used:

142.68 g (Dispersion)

16.12 g (Solid)

16.28 mmoles of Nafion Monomer Units

## Quantity of Water Used:

173.44 g (Actual)

## Resulting Concentrations % (w/w):

5.039 Nafion

0.538 Sodium Persulfate

0.00234 Iron(III) Sulfate

0.263 Baytron M

## Resulting Molar Ratios

2.755 Nafion Acid Functionalities

1.222 Sodium Persulfate

3.170E-03 Iron(III) Sulfate

1 Baytron M

The Baytron - M was added at 755hrs. The reaction mixture was clear & colorless. Experiment is continued on E100074-25.

- \* E100074-23, SE-10072, aqueous Nafion® Dispersion, graciously provided by Allan Cairncross. It is assumed to have an E.W. of 990 & to be 11.3% Solids.
- \*\* Sodium Persulfate, E100046-101, Fluka, 299.0%, Catalog Number: 71899, Lot Number: 398196/1.
- \*\*\* Iron(III) Sulfate, E100048-106, Aldrich, 97%, Catalog Number: 30,7771-8, Lot Number: 055177S.
- The 3-Necked Flask was equipped with a glass stirring paddle powered by an Air-Driven Overhead Stirrer. The Thermocouple has its own inlet. This is a "jacketed" flask, allowing circulation fluid of 22°C to flow through it.
- Test Filtration: Using a syringe the sample was test filtered through a Millipore Millex®-SV 25mm Filter, Catalog Number: SLSVR25LS, Lot Number: ROBN56672, 5.0µm Pore size in succession with a Whatman® 13mm Filter, Poly(Sulfone) Filter Media, Catalog Number: 6780-1310, Lot Number: 99669, 1.0µm Pore size. Syringe used is a 10mL Norm-Ject®, (Lot Number: 00 D 03 8 A; VWR Catalog #: 53546-006; Latex & Silicone Oil Free).
- Results: 1.0µ Filter - Filtrate passed through with a large amount of pressure, then no more sample would pass through the filter, no matter the amount of pressure. 5.0µ Filter - Filtrate passed through with resistance.
- E100061-37, Baytron M, received from Bayer. Lot Number is K0019. Its p is assumed to be 1.334g/mL, as per its Accompanying MSDS.

*Shawn*

EXPERIMENTER

DATE

WITNESSED BY

DATE

E. I. du Pont de Nemours and Company

BOOK PAGE

TITLE \_\_\_\_\_ DATE \_\_\_\_\_

PURPOSE

*Continued from p. 24*

E 100074- 25

*No Writing Underneath!*

At 1110hrs, the reaction mixture was poured into a 500mL Nalgene® Plastic Bottle. Yield is 307.65g. This was then placed onto the roller that resides in the lab of Tom Deubers & Rose Swiatek for several days. This is to be known as E100074-24 until modified or exhausted.

The sample was observed to have a strong smell of Sulfur and VERY foamy. A Test Filtration\*\* was performed.

\*\* Test Filtration: Using a syringe the sample was test filtered through a Millipore Millex®-SV 25mm Filter, Catalog Number: SL5VR25LS, Lot Number: ROBN55672, 5.0µm Pore size. Syringe used is a 10mL Norm-Ject® (Lot Number: 00 D 03 8 A; VWR Catalog #: 53548-006; Latex & Silicone Oil Free).

Results: 5.0µ Filter - Sample did not pass through the filter at all.

*Shawn Yeisby*

EXPERIMENTER

*Shawn Yeisby*

DATE

WITNESSED BY

*Susan L. Hallenbeck*

DATE

TITLE

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PURPOSE

Repeat of 24

E 100074- 35

E100074-35.

Differences: This reaction is conducted like E100074-24 except that DI Water obtained from E928, 3<sup>rd</sup> Floor was used. Reaction will only run for approximately 90 minutes before being split. Nafion Dispersion SE-10072 is used.

Into a corresponding 500mL Nalgene<sup>®</sup> Plastic Bottle SE-10072<sup>®</sup> and the listed amount of DI Water<sup>™</sup> were massed. The Sodium Persulfate<sup>™</sup> and the aliquot of a Stock Solution of Iron(III) Sulfate<sup>™</sup> were then massed into the bottle. The cap was then replaced and the bottle was shaken. The bottle contents were then poured into the Reaction Vessel<sup>™</sup>. (Some losses during transfer had occurred.) The Reaction Mixture was then allowed to stir for 30 minutes prior to addition of the Baytron-M<sup>™</sup>. The reaction mixture had a slight color. The Reaction Mixture temperature was 22.7°C when the Baytron-M was added. The table below shows all of the Quantities of reagents used. This table also shows the results of necessary calculations.

## NECESSARY DATA:

Nafion

990 g/mole of Monomer Unit of Acid

11.3 % Solid Nafion

Sodium Persulfate

238.10 m.w. of Sodium Persulfate

Iron(III) Sulfate

399.87 m.w. of Iron (II) Sulfate

Stock Solution of Iron (III) Sulfate

0.0667 g Iron (III) Sulfate

12.2775 g Diluted to w/ Water

0.5433 mass % of Iron (III) Sulfate

Baytron M

142.17 g/mole

1.334 g/mL

## Amount of Solid Sodium Persulfate Used:

1.72 g

7.224 mmoles

## Amount of Iron(III) Sulfate Used:

1.40 g of Iron(III) Sulfate Solution

0.00761 g of Solid Iron(III) Sulfate

0.0190 mmoles of Iron(III) Sulfate Used:

## Baytron M Addition:

0.830 mL of Baytron M, E100061-37

0.840 g of Baytron M (Calculated)

5.911 mmoles Baytron M (Calculated)

## Mass of Total Reaction Mixture:

319.88 g (Calculated)

## AMOUNTS ACTUALLY USED IN INITIAL REACTION MIXTURE:

## Amount of Nafion Solution Used:

142.68 g (Dispersion)

18.12 g (Solid)

16.29 mmoles of Nafion Monomer Units

## Resulting Concentrations % (w/w):

5.040 Nafion

0.538 Sodium Persulfate

0.00238 Iron(III) Sulfate

0.263 Baytron M

## Quantity of Water Used:

179.45 g (Actual)

## Resulting Molar Ratios

2.755 Nafion Acid Functionalities

1.222 Sodium Persulfate

3.218E-03 Iron(III) Sulfate

1 Baytron M

NO WRITING UNDERNEATH

The Baytron - M was added at 825hrs. The reaction mixture was clear & colorless. Experiment is continued on E100074-36.

- \* E100074-23, SE-10072, aqueous Nafion<sup>®</sup> Dispersion, graciously provided by Allan Cairncross. It is assumed to have an E.W. of 990 & to be 11.3% Solids.
- \*\* DI Water was obtained from E328, 3<sup>rd</sup> Floor on The Vessel that contained the DI Water was carefully rinsed 3 times with the DI Water prior to filling.
- \*\*\* Sodium Persulfate, E100048-101, Fluka, 99.0%, Catalog Number: 71899, Lot Number: 398198/1.
- \*\*\*\* Iron(III) Sulfate, E100048-106, Aldrich, 87%, Catalog Number: 30,7771-8, Lot Number: 05517TS.
- The 3-Necked Flask was equipped with a glass stirring paddle powered by an Air-Driven Overhead Stirrer. The Thermocouple has its own Inlet. This is a "jacketed" flask, allowing circulation fluid of 22°C to flow through it.
- Test Filtration: Using a syringe the sample was test filtered through a Millipore Millex<sup>®</sup>-SV 25mm Filter, Catalog Number: SLSVR25LS, Lot Number: ROBN56672, 5.0µm Pore size in succession with a Whatman<sup>®</sup> 13mm Filter, Poly(Sulfone) Filter Media, Catalog Number: 6780-1310, Lot Number: 99869, 1.0µm Pore size. Syringe used is a 10mL Norm-Ject<sup>®</sup>, (Lot Number: 00 D 03 6 A; VWR Catalog #: 53548-008; Latex & Silicone Oil Free).
- Results: 1.0µ Filter - Filtrate passed through with a large amount of pressure, then no more sample would pass through the filter, no matter the amount of pressure. However, more sample passed through the filter before "infinite" resistance. 5.0µ Filter - Filtrate passed through with some resistance.
- E100061-37, Baytron M, received from Bayer. Lot Number is K0049. Its p is assumed to be 1.334g/mL, as per its Accompanying MSDS.

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E 100074- 36

TITLE

DATE

PURPOSE

Continued from p. 35

Time of Day	Reaction Time, t (hr. min.)	T (°C)	Notes and observations:
<del>8:25</del>	0:00	22.9	Reaction mixture was clear, colorless when the monomer was added. Monomer appeared to be oil droplets dispersed through the reaction mixture.
	1:07	22.7	The Reaction Mixture is VERY Dark Blue.
	1:15	22.7	The reaction mixture was split into a 2-250mL Nalgene® Plastic Bottle. When dismantling the Reaction Vessel, NO gel particles residing were noticed on the stirring shaft. The reaction mixture remained in these bottles until a later time.

**NO WRITING UNDERNEATH**

At the above time of reaction, the reaction mixture was poured into a 2-250mL Nalgene® Plastic Bottle. Yield is 257.10g. Test filtration was not performed. These bottles are to be known as E100074-35 until modified or exhausted.

*Shawn Yeibey*

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WITNESSED BY

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TITLE

PURPOSE

*Resin Treatment of 1/2 of E100074-35*

E 100074- 37

E100074-37,

Half of the sample from E100074-35\* (148.75g) that had remained in a 250mL Nalgene® Plastic Bottle was used for treatment, as suggested by U.S. Patent 5,766,515. This sample was treated with Lewatit® MP 62 WS (7.80g)\*\* & Lewatit® S 100\*\*\* (7.75g). After preparation\*\*\*\* using a plastic powder funnel, the resins were added to the sample in the Plastic Bottle. Using the roller that resides in the lab of Tom Deuber & Rose Swiatek, the resulting slurry was rolled from 945hours ~~2000/02/12~~ to 900hours of ~~2000/02/12~~.

**NO WRITING UNDERNEATH**

Separation of the slurry was done by suction filtration through a Coarse Fritted Funnel. Suction was then applied to facilitate filtration. Care was used to ensure no loss of solvent. Yield is 110.21g. The resulting product is known as E100074-37 until modified or exhausted. Test Filtration<sup>5\*</sup> Results - 5.0µ: Sample passed through with no change in color and with little difficulty. After a while the sample "plugged" the filter. 1.0µ: No filtration whatsoever.

\* E100074-35, Polymerization of Baytron M with Nafion Dispersion SE-10072 (E100074-23).

\*\* E100061-25, Catalog Number: NM03, Lot Number: H0332.

\*\*\* E100048-127, Catalog Number: NA01, Lot Number: 06Y71176.

\*\*\*\* All mentions of DI Water were obtained from the 3<sup>rd</sup> DI Water Tap in E328. Each of the resins was massed into a 1000mL Beaker. DI Water was then added to resin mixture. The liquid phase was poured away from the resin. Fresh DI Water was then added. This process of washing was repeated until there was no qualitative trace of impurities, i.e. no color or surfactant action. The slurries were combined then suction filtered through a Coarse Fritted Glass Funnel. Using a Spatula, the resins were then transferred to the bottle that contained the sample. Care was taken not to take the resins to "dryness". Care was also taken not to scratch the Glass Frit. The numbers in parentheses are the masses of the resins Pre-Treatment. Obviously some losses due to transfers had occurred.

<sup>5\*</sup> Test Filtration. Using a syringe the sample was test filtered through a Millex®-SV 25mm Filter, Catalog Number: SLSVR25LS, Lot Number: ROBN56672, 5.0µm Pore size in succession with a Whatman® 13mm Filter, Poly(Sulfone) Filter Media, Catalog Number: 6780-1310, Lot Number: 99669, 1.0µm Pore size. Syringe used is a 10mL Norm-Ject®, (Lot Number: 00 D 03 8 A; VWR Catalog #: 53548-006; Latex & Silicone Oil Free).

*Shawn Yeisley*

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DATE

## E. I. du Pont de Nemours and Company

BOOK PAGE

TITLE Co-Final DATE \_\_\_\_\_

PURPOSE \_\_\_\_\_

E 104328- 51

148.75 g of the aqueous PEDOT/Nafion® in one of the two plastic bottles was further treated with two ionic exchange resins. One of the two resins is Lewatit® S100, a trade name from Bayer, Pittsburgh, PA, USA for sodium sulfonate of crosslinked polystyrene. The other ionic exchange resin is Lewatit® MP62 WS, a trade from Bayer, Pittsburgh, PA, USA for free base/chloride of tertiary/quaternary amine of crosslinked polystyrene. The two resins were washed before use with deionized water separately until there was no color in the water. 53 g of Lewatit® S100 and 51 g of Lewatit® MP62® WS were then mixed with the 158 g aqueous PEDOT/Nafion® dispersion in a plastic bottle. The bottle was then placed on a roller for stirring for 23 hours. The resulting slurry was then suction-filtered through a coarse fritted-glass funnel. Yield is 110.2 g. Based on elemental analysis of the sample dried from a 2.6 % (w/w) dispersion, it contains 21.75% carbon, 0.23 % hydrogen, 1.06 % nitrogen and 2.45 % sulfur. Other elements such as oxygen and fluorine were not analyzed. To remove fluorine interference with sulfur analysis,  $\text{CeCl}_3$  and a cation exchange resin was added.

10 g of the PEDOT/Nafion® dispersion was mixed with 10.01 g deionized water, which constitutes 2.6 % (w/w) solid based on a gravimetric analysis of dried solid. The aqueous PEDOT/Nafion® dispersion was then tested for light emission properties. The glass/ITO substrates (30mmx30mm) having ITO thickness of 100 to 150 nm and 15mmx20mm ITO area for light emission were cleaned and subsequently treated with oxygen plasma. The aqueous PEDOT/Nafion® dispersion was spin-coated onto the ITO/glass substrates at a spinning speed of 700 rpm to yield 96 nm thickness. The PEDOT/Nafion® coated ITO/glass substrates were dried in air at 200°C for 3 minutes. The PEDOT/PSSA layer was then top-coated with a super-yellow emitter (PDY 131), which is a poly(substituted-phenylene vinylene) from Covion Company (Frankfurt, Germany). The thickness of the EL layer was approximately 70 nm. All film thickness was measured with a TENCOR 500 Surface Profiler. For the cathode, Ba and Al layers were vapor deposited on top of the EL layers under a vacuum of  $1 \times 10^{-6}$  torr. The final thickness of the Ba layer was 30 Å; the thickness of the Al layer was 3000 Å. Device performance was tested as follows. Current vs. voltage curve, light emission intensity vs. voltage curve, and efficiency were measured with a Keithley 236 source-measure unit from Keithley Instrument Inc. (Cleveland, OH), and a S370 Optometer with a calibrated silicon photodiode from UDT Sensor, Inc. (Hawthorne, CA). Five measured light emitting devices show operating voltage ranging from 3.2 to 3.3 volt and light emission efficiency ranging from 8.3 to 9.8 Cd/A at 200 Cd/m<sup>2</sup> brightness. They have stress half-life ranging from 243 to 303 hr at 80 °C.

Continued next pageEXPERIMENTER Chen et al

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